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FOOD PROCESSING WITH ELECTRONS

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The prime facts are these. Electrons can kill microorganisms and insects by a quick, almost heatless process, yet even very energetic electrons can penetrate a contaminated material only a short distance to do the killing. An inch and a half of water or vegetable or animal tissue or other organic material is at present about the practical thickness limit for electron sterilization.

Electron sterilization is largely a postwar venture, and is now at an advanced laboratory stage. Almost the entire development has come from four places: Massachusetts Institute of Technology, Cambridge, Massachusetts, Electronized Chemicals, Inc., Brooklyn, the General Electric Research Laboratory, Schenectady, and High Voltage Engineering Corp., Cambridge, Massachusetts. To most of us the idea that electrons may flow through wires or vacuum tubes is common enough to be accepted, yet the notion that electrons in quantity may also be made to stream into the air and there, impinging on a foodstuff or a drug, free it from organisms causing spoilage or infection, seems bizarre.

The apparatus that produces high energy electrons being expensive and electron penetrating power being severely limited, it may be guessed that electron sterilization will be tried commercially first on commodities with high unit value that are normally packaged in small containers - biologicals, for example. Preserving food is a lot more difficult. Allowable costs are low and the thickness of material one would like to electron sterilize is usually much greater than can be so sterilized. Besides microorganisms, which

the electrons will destroy, there are usually enzymes that must be inhibited somehow if the food is to keep its quality. Nutritive value, flavor, texture and color have to be retained. And then, toxic or growth-retarding substances possibly created by the electron irradiation have to be looked for through prolonged animal feeding experiments.

Among primarily agricultural applications of electron irradiation, food processing would seem to be so important that most of what follows will be devoted to it. Other conceivable large scale applications - preservation of stored grain and oilseeds, and treatment of quarantined plants are examples - will be described at less length in the thought that commercialization, if it comes about at all, will develop only after the method has been a success when applied to more expensive or less bulky commodities.

IONIZING RADIATIONS

Sterilizing electrons (called also cathode rays) are given millions of volts energy solely that they may have appreciable penetrating power. As the electrons traverse matter they ionize it, losing on the average 30 electron volts (ev) for each ion pair they create. A 3 million volt (3 Mev) electron will, then, by the end of its path have created 100 thousand ion pairs. When the ionization takes place in a microorganism or an insect the body chemistry may be sufficiently altered that the organism dies. Inevitably the chance that a single ionization will be lethal varies with the species and strain of organism and its physiological state. To relate kill with total ionization produced in the irradiated specimen the dosage unit in vogue is the "rep." This derives from the jargon of X-ray and radium therapy. It is the amount of radiation that produces 1.6×10^{12} ion pairs per gram of absorber and thereby dissipates 93 ergs of energy. A million rep is a sterilizing dosage for all but the most resistant microorganisms in their radiation-resistant spore form; expressed as heat it is equivalent to about 2 calories, so that the maximum temperature

rise in a material composed mostly of water is 2 deg. C., or, in a dry organic material, about 5-6 deg. C. As a result of a million rep dosage only one atom per 30,000 in the absorbing material is ionized.

Possible competitors for electrons in heatless sterilization are X-rays and gamma-rays. Both are photons, X-rays man-made and gamma-rays spontaneously emitted by radioactive materials. Photons sterilize by the same ionization mechanism as electrons. The probability of ionization by photons is, however, much less, with the result that photons effectively penetrate 50-100 times as far. The reason we are concerned with electron rather than photon sterilization is simply the low power of photon sources. It may not always be thus, for potentially uranium fission by-products can provide photon dosages comparable to the electron dosages now possible from commercial electron accelerators - a billion or more rep per second. The Atomic Energy Commission through its Stanford Research Institute Project 361 is trying to find out how feasible photon sterilization is. The Atomic Energy Commission is sponsoring food preservation experiments with gamm-rays from radioisotopes at Columbia University, Massachusetts Institute of Technology, and the University of Michigan. In these experiments the cobalt-60 gamma-ray sources are by usual standards very strong, equivalent to a kilogram of radium. Yet so disparate is the power from photon and electron sources that a sterilization accomplished by electrons in a fraction of a second takes the kilocurie gamma-ray source four hours. X-ray sterilization apparently can be dismissed because X-ray generator efficiency is so low. Whichever, if either, kind of ionizing "radiation" is ultimately adapted for food preservation, the experience gained with both photons and electrons in the present developmental phase is bound to be useful.

ELECTRON ACCELERATORS

Today, there are three types of accelerators that produce 1 to 5.5 Mev electrons in large enough quantity to do research on the preservation of foods: High Voltage Engineering Corporation's Van de Graaff generator; General Electric Company's resonance transformer accelerator; and Electronized Chemicals Corporation's capacitron. None is scaled for commercial operation.

Van de Graaff Electrostatic Accelerator. The Van de Graaff apparatus builds up its potential by mechanically transporting electrons on a cotton belt from near ground potential to a collecting element which is really one plate of a simple electrical condenser. The greater the quantity of electricity thus transferred, the greater the potential difference between the terminals at the upper and lower ends of the belt, and the greater the energy which can be given to electrons in an auxiliary accelerating tube. In the 2 million volt model the belt is 6 inches wide and travels 4,000 feet a minute around pulleys about 4 feet apart. A 30 kilovolt rectifier unit sprays electrons on the belt. A metal-insulator laminated tube provides uniform gradient from the high-voltage terminal to ground potential. Loss of charge to the atmosphere is prevented by operating the entire assembly in a tank filled with dry nitrogen-carbon dioxide at a pressure of 350 p.s.i. Insulation provided in this way makes possible a compact apparatus 3 feet in diameter and 5-1/2 feet high. Parallel to the belt and likewise inside the laminated supporting tube is the accelerator tube. This, which also is laminated for uniform potential gradient, is evacuated continuously by a diffusion pump. Electrons emitted from a hot cathode at the upper end of the tube are accelerated by the high negative potential generated as described and pass through an aluminum foil window at the bottom of the tube into the air. Potential can be varied between 0.75 and 2.0 million volts. The maximum current, 100 microamperes, is imposed by failure of the window through overheating.

Operated as an X-ray tube, with a gold target rather than an aluminum foil in the beam, the allowable current is 250 microamperes. At 100 microamperes and 2 million volts the power output of cathode rays is 200 watts. This corresponds to about 20 million rep per second, or to a practically sterilizing dosage of 1 million rep applied to 20 grams of a foodstuff per second. About 20 of these accelerators have been sold at a price of roughly $75,000 each. Van de Graaff generators characteristically produce a continuous current of electrons, all of which have the same energy. The most apparent weakness of this generator is the belt, which must be replaced periodically, and which in higher power-installations must be made wider in proportion to the increased current drawn from the accelerator tube. The largest commercial Van de Graaff produces 3.3 milliamperes at 5.5 Mv. Its power, 18 kilowatts, if fully utilizable, could give a million rep dosage to almost 2 kilograms of material a second.

Resonance Transformer. A resonance transformer is so constructed that the natural period of electrical oscillation of its secondary winding is equal to the frequency of the exciting current. It is thus able to operate with fair efficiency without an iron core. Elimination of the core makes insulation much simpler and also makes it possible to place an X-ray or electron-ray tube coaxially with the windings. During the last dozen years, General Electric Company has sold over a hundred of these outfits with 1 Mev peak energy for industrial radiography and X-ray therapy. The price of the 1 Mv X-ray unit is $50,000 to $75,000 (installed), and for the similar 2 Mv unit it is about $125,000. The million volt transformer operates at 180 cycles per second. Its output is rectified in the accelerating tube, and current is actually drawn only when the voltage is between 85 and 100% of the peak. The accelerated electrons, then, are emitted in a succession of short bursts which recur 180 times a second. A mean current of 10 milliamperes can be taken from the transformer. The corresponding power is about 9 kilowatts, and, as electron rays,

is equivalent to a billion rep per second. When operated as an X-ray generator, with a tungsten target in a water-cooled copper mounting placed in the electron beam, the largest current that can be drawn continuously is 3 milliamperes.

The only apparatus at present powered by a resonance transformer and operated as an electron generator is an 800 kilovolt peak apparatus in the General Electric Research Laboratory. This is operated very conservatively, at 100 microamperes, to avoid overheating the 3 mil stainless steel window in its sealed-off acceleration tube. It is reasonable to suppose that the current could be raised to several milliamperes if a much larger window, say a foot in diameter, were used, and if frequent window replacement and continuous evacuation of the tube were made standard operating procedure. Happily, an electron beam can be expanded or contracted pretty much at will by means of magnets. The General Electric resonance transformers are insulated with Freon-12 at a pressure of 60 p.s.i. The 1 Mv apparatus is enclosed in a tank whose dimensions are about the same as the 2 Mv Van de Graaff accelerator, whereas the tank for the 2 Mv resonance transformer is 5 x 8 feet. It is not known whether General Electric regards it as feasible to build resonance transformer and accelerator tubes for service at much more than 2 Mv. In computing the number of rep--or better the amount of material that can be sterilized per second--from the current and voltage, an allowance of 20% or more must usually be made for electron absorption by the grid that supports the window and by the several inches of air that usually lies between the window and the objects to be irradiated, and particularly for inhomogeneity in the electron beam.

Impulse Generator. Electronized Chemicals Corporation's electron accelerator is an impulse generator which it calls a capacitron. It consists of a group of, say, 30 condensers charged to 100,000 volts each, and then discharged in series so that the maximum voltage attained is 3 million. The impulse generator characteristically produces a heavy current at a high voltage

for a short time. It is a sort of artificial lightning generator and has been used as such for 20 years by manufacturers of electrical transmission equipment. The capacitron discharge takes place in an evacuated laminar tube 3 feet high and 6 inches in diameter inside. A burst of gas introduced near the upper end when the condensers are charged provides electrons for the current of several thousand amperes, which flows for about a millionth of a second. The electron stream issues from the tube through a 0.05 mm. thick aluminum window at the bottom. The capacitron now in operation is rated at 3.5 million volts, but is actually used at 2.2 million volts. This is the peak voltage, and the mean seems to be nearer 1.75 million volts. The radiation output per impulse is stated to be 200 million rep. The cycle of operations takes about 10 seconds. The capacitron to date has been very definitely a laboratory apparatus. Electronized Chemicals Corporation has had for 4 years a design for a "commercial prototype" capacitron, with a 10-inch window, a peak voltage of 5 million, a discharge rate of 50 to 100 cycles a minute, and an output of a billion or more rep per discharge. This apparatus is still only partly built.

Other Accelerators. For higher energy and greater penetrating ability electrons may be accelerated in a betatron or a synchrotron. Allis-Chalmers has sold a dozen or more 24 Mev betatrons, at about $90,000 each, and General Electric offers a 10 to 70 Mev synchrotron for about $250,000. For their present use in radiation therapy and in nuclear research, the small electron current of the accelerators is acceptable. Reportedly, there is a very good prospect that their power output can be vastly increased if needed in some application such as sterilization of biological material.

In a betatron, electrons are accelerated in an evacuated glass tube, shaped like a doughnut and placed between the poles of a magnet, which may weigh from 1 to 100 tons. The magnet is powered by alternating current.

During the quarter cycle when the magnetic field rises from zero to its maximum value, the electrons move in a stable circular orbit, picking up energy continuously from the magnetic field at the rate of perhaps 100 electron volts per revolution. At the end of the quarter cycle they may have traveled 100,000 times around the orbit and, without application of large voltages, have acquired energy equivalent to electrons accelerated by a potential difference of 10 million volts. They can then be directed through a window or on a target to produce X-rays, as desired.

The electron synchrotron first accelerates electrons to about 2 Mev energy, thereby giving them 97% of the velocity of light, by the betatron principle. Then, while a steady magnetic field holds them in a stable orbit, they are accelerated to the desired final energy by radio-frequency power applied across a gap between silver-coated segments of the inner surface of the glass tube. The linear electron accelerator, which is not now a commercial apparatus, omits the magnet of the synchrotron and, instead of accelerating repeatedly across a single gap, accelerates by applying r-f to a sucdession of gaps between tubes in end-to-end array.

From the above, it will be noted that all generators, with the exception of the impulse generator, release energy as a practically continuous stream of electrons. The effect on material treated by these two methods is said to be entirely different, but the evidence presented has not been convincing. For instance, it has been reported that ergosterol treated with a sudden burst of electrons (impulses of the order of 10^{-6} sec.) did not bring about the formation of vitamin D_2, but that continuous radiation with alpha or beta particles did cause vitamin D_2 formation (1). Again, casein and egg albumen irradiated by impulses caused no change in the material, while continuous radiation with beta or alpha particles brought about decomposition and oxidation. However,

data obtained from modern high power continuous current machines do not substantiate such differences.

INDUSTRIAL HAZARDS OF PROCESSING WITH HIGH ENERGY ELECTRONS

No radioactivity is produced by electron sterilization, either in the material sterilized, its container, or in any associated handling equipment. The sole hazard arises from the electrons themselves and especially from the X-rays that are always associated with them. The sterilization dosage for foodstuffs is 1 to 2 million rep. The lethal dosage for a human is only about 500 rep. This is for "whole body exposure," and the dosage is apparently nearly the same for X-rays, which cause tissue damage throughout the entire body volume, and for electrons, whose effect is confined to tissues near the surface. According to present ideas, the highest permissible exposure for those who work regularly with X- or electron-rays is 0.3 rep per week. It is, therefore, obvious that the rays must be almost completely attenuated by absorbers or by distance at all sites where operators or others might be exposed to them.

If electrons from the generator were absorbed only in organic material there would be no particular hazard, because the X-rays produced would be low in intensity and penetrating power. A greater hazard arises from penetrating X-rays that originate when cathode rays are stopped by material of high atomic number, such as steel or brass in the electron accelerator itself, or in auxiliary equipment for manipulating specimens for irradiation. In a laboratory installation that produces a 1 milliampere beam of 3 Mev electrons the shielding requirement might be met in this way: a stack of 4-inch lead bricks surrounding the irradiation site - the accelerator tube adjacent to the exit window, the specimen, and the specimen support or conveyor; 12-inch concrete walls and ceiling in the irradiation room; and a 15-foot minimum distance of approach to the target during irradiation. A commercial electron sterilizer or a laboratory

apparatus that produces a milliampere or so of 20 Mev electrons might better have its irradiation chamber several feet underground. In all installations access doors and other hazardous areas would have electrical interlocks to prevent operation of the accelerators when the hazardous areas are occupied. In addition, there would be area monitoring for X-radiation, and personal monitoring of operators by pocket ionization chambers, film badges and periodic blood testing as is done routinely in radioisotope experimentation.

PENETRATION AND ENERGY

The depth to which electron rays penetrate a material is proportional to their energy. In substances of density 1, which is a good enough approximation for foodstuffs, the depth of greatest penetration is 4.5 mm. for each million electron volts energy. Only an infinitesimal proportion of electrons attain this depth, though, and the effective depth for sterilization is nearer 3 mm. per Mev. On a scale of 100, the bacteria-killing power of 1 Mev electrons increases from 60 at the surface to 100 at 1.4 mm., then decreases to 55 at 3 mm., to 11 at 4 mm., and to 0 at 4.5 mm. It follows that if a lethal dosage is provided at 4 mm., the zone near 1.4 mm. will have received 9 times the lethal dosage, and everything deeper than 4 mm. will be unsterile.

The most effective use of electrons is made by irradiating the food in a flat container from opposite sides. By doing this the depth at which sterility is assured is more than doubled because use is made of the overlapping "tails" in the graph of killing power versus depth (2). To illustrate with 3 Mev electrons, if a sterilizing dosage of 60 is supplied at the top surface, the dosage rises to 100 at 5 mm., falls to 60 at 12 mm., rises again to 100 at 20 mm., and falls to 60 at the bottom surface at 24 mm. Apparently, so long as the primary electrons have equal energy, substantially more than the sterilizing dosage must be given to most of the material irradiated in

order that all of it shall have at least the sterilizing dosage.

Penetration by electrons varies inversely as the density of the material irradiated. Thus, the steel wall of a tin can (0.26 mm. thick, and density 7.8) absorbs as much energy from the electron beam as 2.6 mm. of water or tissue, and in effect reduces the sterilizing beam energy by about 3/4 Mev. An aluminum wall of the same thickness is equivalent to 0.9 mm. of tissue; and a glass bottle wall 2.6 mm. thick is equivalent to 7 mm. of tissue. It is clear from this that the container for food during electron sterilization should be made from a light thin material such as cellophane or polyethylene, with subsequent packaging in a sturdier container if necessary. Air, because of its low density, is reasonably transparent to cathode rays, and transmission of 3 Mev rays through 6 inches of air amounts to 90-95%.

Five and one-half Mev electrons are the most energetic now available at high intensity from commercial generators. This places a strict upper limit of about an inch and a half to the thickness of foodstuff that may be electron-sterilized. If irradiation is from one side only, the greatest thickness is little more than half an inch. Electron sterilization of food packages of thickness comparable to the diameter of a No. 2 can, 3-7/16 inches, must wait upon the development of higher energy electron accelerators. A linear accelerator or synchrotron with 1 milliampere output at 20 Mev would be ideal for experimental purposes. Such has not yet been built.

EFFECTS OF HIGH ENERGY ELECTRONS

Historical:

Reports that cathode rays kill bacteria were made as early as 1914 (5) and later by Coolidge in 1925 (6), by Wychoff and Rivers in 1930 (7), and by Brasch in 1933 (8). Up to 1947 the principal experiments with high energy electrons outside the generating tube were performed in the General Electric Research Laboratory by Coolidge and Moore. Apparently most of the experiments

were done about 1924-25, and were described in several almost equivalent publications between 1925 and 1932 (6, 9, 10, 11). Development of the high voltage electron tube was Coolidge's chief accomplishment and he expected that its major use would be to generate X-rays for medical and industrial radiography. His experiments with Moore ranged widely and were correspondingly cursory. Coolidge and Moore observed: luminescence and scintillation in crystals; canalization in minerals, gums, organic acids, alcohols and celluloid at -180 deg. C.; aqueous solutions of sugar, starch and glycerine became acid on irradiation; milk and butter quickly acquired a rancid flavor; Staphylococcus aureus, Escherichia coli, Bacillus prodigiosus and Bacillus subtilis were destroyed; fruit flies, water snails and cockroaches were killed; cells of Ficus elastica became permeable to latex; changes in the tissue of a rabbit's ear ranged from erythema to destruction that required amputation; vitamin D_2 was produced from ergosterol; drying rate, color, refractive index and molecular weight of linseed and china-wood oil were changed; castor oil polymerized to a solid; a variety of condensation and decomposition products, almost none of them identified, were obtained from acetaldehyde, ethylene, ethanol, acetone, and acetylene.

Effect on Foods:

The first significant publication relating to high energy electron preservation of foods was by Brasch and Huber in 1947 (3). They used an impulse generator similar to one built by Brasch about 1930 with financial support from the Allgemeinen Elektricitäts-Gesellschaft (4). Considerable research has also been undertaken by Huber, Brasch and their associates in the Electronized Chemicals Corporation on the effect of high-speed electrons on drugs, viruses and bacteriophages, but these will not be discussed here.

Several foods have been treated by means of the capacitron at impulses of 10^{-6} sec. (1). These foods were packed in glass, plastic or metal foil, and stored at room temperature from 2 to 264 days. Reps used were not stated. Butter, margarine and cream cheese assumed an off-taste in about 90 days; boiled ham decomposed slightly in 63 days; roast beef darkened in 91 days; peas, beans and carrots bleached in 184 days; diced potatoes browned in 64 days; broccoli showed some bleaching in 42 days; lettuce was soggy and bleached in 2 days; orange juice lost some of its aroma in 128 days. In other foods, such as sweet cherries (64 days); grapefruit juice (134 days); sliced pineapple (94 days); sliced coconut (86 days); lima beans (227 days); hamburger (83 days); bacon (156 days); flounder (127 days), and several other meats, there was no change. There are no data as to how these different foods were prepared or how critically they were judged; it is not known whether the vegetables were blanched.

Ultra-short-time impulses may bring about taste changes (called "irradiated taste") in some cases. To eliminate these changes, or at least to keep them to a minimum, the importance of having optimum outside conditions during irradiation is emphasized. These optimum conditions may be cooling or partial evacuation, or both (12), or, perhaps, by the addition of chemical compounds as free radical acceptors (2). Proctor, in his work on fish, irradiated with from 900,000 to 5,700,000 rep by means of a Van de Graaff apparatus, mentions a "cooked" and "oxidized odor resembling ozone" and "strong smell of ozone" (13). The higher the dosage, the more pronounced the ozone odor. Proctor and O'Meara (14) sterilized milk with 3×10^{6} rep at 50-60 deg. F. and reported off-flavor and off-odor production, which decreased during storage. These changes were minimized by irradiating the milk in the frozen state, but a larger dosage for sterilization was required. These same workers reported off-flavor when orange juice was irradiated.

Proctor and his co-workers have treated certain dried fruits with dosages up to 10 million rep (15). Bleaching of the flesh of prunes was observed immediately after treatment, but there was no color change in the skin. After 2 weeks at room temperature, the flesh returned to normal color. There was no change in flavor immediately after irradiation or 2 weeks later. The skin and flesh of Thompson Seedless raisins were bleached and there was a definite off-taste. Color returned to normal after 2 weeks. Prune and apricot juices were bleached, but the color partially returned in 2 weeks. Hamburger and canned chopped meat products have been treated with cathode rays (14). In some instances there was discoloration of the meat, and generally off-flavor development which was dissipated when the meat was cooked. A slight increase in the peroxide number of the fat in hamburger was noted after irradiation.

Effect on Enzymes. The effect of high intensity electron bursts on enzymes varies considerably. With 100% sterilizing doses many enzyme systems in tissues remain intact, without giving rise to autolyses, even after long storage periods at room temperature. For instance, doses of electrons sufficient to give complete bacteriological sterility cause little or no inactivation of proteolytic enzymes in tissues (16). Amylase subjected to an electron burst of 1×10^6 rep lost 5.5% of its activity; an electron burst of 1.5×10^6 rep caused only a 14% loss in lipase activity; diastase with a dose of 6×10^6 rep suffered a 31% loss in activity, while hyaluronidase with a dose of 4×10^6 rep suffered a 76% loss of activity (8). The preparations were in powder form in order to eliminate the effect of activated solvent molecules.

Dunn and his co-workers (17) irradiated milk with electrons, but they apparently made no observations on the keeping qualities of the milk. Cottage cheese was made from the milk and "there appeared to be no important organoleptic

differences between the cheeses produced from the raw and cathode ray-treated milks." Proctor and O'Meara (14) found that it required a greater dosage to destroy phosphatase in raw milk than to destroy peroxidase, and, in the case of apple juice, a dose of 1 million rep did not destroy phosphatase.

Inasmuch as proteolytic enzymes are not inactivated by a sterilizing dosage of radiation, how can it be explained that no autolysis was observed in meat preserved by this method? It is postulated (8) that either inactivation has taken place at a point of the enzymatic chain which is not revealed by the testing technique used (which is unlikely), or that enzymatic breakdown may have to take place with the assistance of microorganisms. In this connection, it has been postulated that autolysis in fish may be due to bacterial action and not enzymatic action (14).

It apparently requires greater dosages of electrons to destroy enzymes in frozen material than in the same material in the unfrozen state (14).

It would not always be necessary to inactivate enzymes by electrons and, perhaps, it would not be economically feasible to do so, because high dosages are required. Inactivation could be carried out by means of conventional blanching methods in the case of vegetables to be canned or frozen.

Effect on Vitamins. The effect of irradiation by electrons on vitamins has been studied by Proctor and his co-workers, and by the Electronized Chemicals Corporation. For the most part, Proctor used pure solutions of the vitamins and found that B_2, C, niacin, and carotene were sensitive to electrons, destruction being greater the greater the dilution (18). In some instances, destruction was less when several vitamins were present together, or in the presence of other substances. There was less destruction of niacin, for instance, in the presence of methionine (19).

On the other hand, Huber points out in his work with polyvitamin preparations that the radiation sensitivity of a given vitamin is a property

which is characteristic of the compound itself, and is not dependent on its mixture with other vitamins (8).

It may be that the destruction of vitamins found by Proctor was due to the effect of activated solvent molecules, since his vitamins were irradiated in pure solution. Indirect radiation effect caused by activated solvent molecules is said to be negligible as soon as inert acceptor molecules are present in sufficient quantities (20). Huber (8) did not find significant losses of thiamin, riboflavin, niacin, ascorbic acid, pyridoxine, vitamin A, and vitamin D when these were irradiated in situ.

That there is a relation of the radiation effect to the composition of the medium has been pointed out by Huber (8). In a report by Huber et al (20), aqueous solutions of ascorbic acid and an orange juice concentrate were treated with 500,000 to 5,000,000 rep. It is assumed that concentration of ascorbic acid was the same in the aqueous solutions as in the orange juice concentrate, although the abstract is not quite clear on this point. A dose of 800,000 rep, which was sufficient for 100% sterilization, caused a loss of 40% of the vitamin in the aqueous ascorbic acid, while there was only a 5% loss of vitamin C in the juice concentrate. Similar results have been obtained by Proctor and O'Meara (14). Thus, it is believed that the destruction of vitamin C in aqueous solutions is primarily an artifact due to the action of activated solvent molecules, and greater retention in orange juice is due to the presence of a heterogeneous group of substances probably acting as protecting substances.

It has been pointed out (8) that destruction of vitamins by radiation is less pronounced at low temperatures than at higher temperatures. It is postulated that this shows that the action of electrons, particularly in aqueous solutions, at room temperatures is an indirect one which can be exerted over rather considerable distances by movement of ions, radicals or by energy transfer

between colliding molecules. In a frozen system mobility is considerably reduced, and there is, therefore, a decrease of the indirect effects of radiation. Proctor and Goldblith (19) noted that the destruction of niacin by cathode rays was indirect, but offered no further explanation.

The indirect effect of electron irradiation on vitamins, enzymes and microorganisms would have an important bearing on the commercial application of this method of preservation. It would seem, however, that much further research will have to be done in order to discover and understand the optimum conditions for treating different types of foods.

Effect on Amino Acids. In addition to vitamins and enzymes, amino acids form another group of compounds of importance. Proctor and Bhatia (13) irradiated haddock fillets with cathode rays produced by a Van de Graaff generator. Haddock was selected as the typical protein, because it is rich in all the "essential" amino acids. Assays were made before and after irradiation with 900,000, 2,700,000 and 5,700,000 rep of ten amino acids (arginine, histidine, leucine, lysine, methionine, phenylalanine, threonine, tryptophane, valine, and cystine). No significant destruction of any one of these amino acids was noticed as the result of irradiation in the doses mentioned above. Proctor and Goldblith (14) also irradiated ground beef patties and protein hydrolyzates with 1.5 million rep. Losses in essential amino acids were small.

Effect on Microorganisms. Both Dunn and his co-workers, as well as Brasch, have studied this phase of the question, but most of the published information has been put out by the latter. The published work by Dunn and his co-workers has been limited to five organisms in pure cultures (17).

Brasch et al (21) studied the effect of penetrating electrons on a variety of bacteria and found that the sterilizing effect is not limited to any

particular species. The amount of radiation needed to achieve 100% sterilization depends roughly upon the size of the organism as well as upon its specific radiation resistance. Vegetative bacteria are most radiation sensitive, whereas small viruses are most radiation resistant. Spores have radiation resistivity, and it is postulated that this may be due to the fact that for inactivation a multiple ionization has to occur in the target (8).

Huber (8) states that in the killing of microorganisms, the numbers killed by successive increments of electron dose are not equal, but each increment of dose kills the same proportion of the number of organisms which have survived until then. Thus, the survival curve is exponential. This should be true according to the so-called "target theory." This theory is based on the premise that ionization must occur within the region where the initial biological effect takes place. This region is referred to as the target and it is necessary for an ionizing particle to pass through it. A hit is defined as ionization within the target caused by a particle (electron) passing through the target. The great majority of biological effects caused by corpuscular radiation are alleged to be of the single hit type, e.g., the finite effect is obtained by a single ionization within the target. In this connection, Proctor and Goldblith (14) have recently found that the electron beam of a continuous electrostatic accelerator can be made more uniform by scattering the electrons with aluminum foil. Therefore, some of the lethal dosages reported may be higher than actually required at present.

In most of the published work on the effect of electrons on bacteria, the organisms were pure cultures in culture media. Data concerning the effect on bacteria and spores in situ are limited. There is a report of 99.9% destruction of <u>Serratia</u> <u>marcescens</u> in pork meat patty and 93.3% destruction of <u>Bacillus</u> <u>subtilis</u> in potato patty (3). No data are given of the initial

bacterial population and without reference to reps used, percentage destruction given is meaningless. The data are not too helpful from another point of view. Both organisms are aerobes (B. subtilis is a facultative aerobe) and S. marcescens does not form spores. Of much more importance would be a study of the destruction of Clostridium botulinum and its spores in known concentrations when added to foods, buffer solutions or nutrient media contained in the consumer package. The study should be extended to certain other putrefactive anaerobes whose spores are more resistant than those of Cl. botulinum.

THE ROAD AHEAD

From what has already been said, it would appear that preservation of foods by electron irradiation may have commercial possibilities. But, before commercial application can be realized, considerable more research must be undertaken, both from the economic standpoint as well as from the standpoint of quality of the product, possible harmful residues, changes in constituents of foods that may affect their nutritional value, and public health implications of foods so treated.

It has already been shown that irradiation of certain foods by electrons may destroy insignificant amounts of vitamins in those foods. However, the data are limited and further researches are necessary.

Published data on animal feeding tests of irradiated foods are meager. Proctor and Goldblith (14) fed a limited number of rats over a period of two years with irradiated foods. No toxic effects were observed and the animals had as long a life span as the controls. However, results of feeding experiments covering several generations have not been published and it is quite possible that unfavorable results would be apparent only after several generations.

Irradiation of brine or water packs of vegetables would bring about a certain degree of ionization of the water with the formation of peroxides.

It is not known whether these peroxides would cause flavor changes in such packs. Distilled and tap water irradiated with dosages comparable to those causing complete sterilization formed hydrogen peroxide in amounts less than .005% (3). We do not know how much peroxide is formed in foods during irradiation.

There appear to be no detailed published data on long-time controlled storage of food products preserved by irradiation and those preserved by conventional methods. It must be recognized that published claims of prolonged quality retention at room temperature have not been substantiated; the claims seem extravagant and the experiments on which they are based have not been fully described.

There are color changes brought about by irradiation of some foods. In the case of prune juice and apricot juice, it has been suggested that this results from the destruction of 5 (hydroxymethyl) furfuraldehyde. Obviously, such changes are important to consumer acceptance. Sometimes these color changes are merely transitory, and sometimes color changes may be inhibited by irradiation at low temperatures.

There are apparently flavor changes which have been described as "irradiation taste" and "ozone" odor. These changes, too, are important to consumer acceptance. In some instances, these flavors are eliminated when the food is cooked.

It would also be of value to determine if electrons follow preferred paths in foodstuffs as they seem to in plastics and brittle crystalline materials.

Nothing has been said relative to the economic aspects of processing with high energy electrons. The published data regarding costs of installation, maintenance and processing are meager (22). There is far too little data on the dosages actually required to sterilize; lacking these, cost estimates are accordingly in error.

It is axiomatic that research on radiation sterilization carried on in the Department or elsewhere under the Department's auspices should be performed with the utmost objectivity and publicized with proper reserve. Considering the startling and often controversial chemical and biological effects observed with high energy irradiation, collaborative tests between laboratories engaged in this type of research should be encouraged. Unfortunately it appears such tests have been lacking to date.

POSSIBLE APPLICATION OF HIGH ENERGY ELECTRONS TO SOME AGRICULTURAL COMMODITIES

As pointed out above, certain fundamental data must first be obtained before this method of treating food materials can safely be applied to agricultural products. Discussed below are some problems where cathode ray treatment could be of value. It is quite probable that machines of, say, 20 Mv could not be economically installed in populated areas because the expense of protection to exclude harmful radiation would be prohibitive.

Cereals and Seeds. Wheat production in the United States amounts to over a billion bushels annually. Stored grain losses caused by insects in the United States alone have been stated to total approximately 6,000,000 bushels a year (22a).

A figure of $600,000,000 has been given as the value of grain (barley, wheat, corn and soybeans) destroyed each year by insects (23), with the implication that most of this could be saved by cathode ray treatment. This figure is misleading because in 1949 nearly 80% of the corn, 40% of the barley, and about 6% of the soybean production never left the farm where cathode ray treatment would be impracticable. Wheat is now fumigated to destroy insects, the cost of such treatment amounting to about 1/2-cent a bushel. If electron treatment can compete with this figure, it would, perhaps, be better than fumigation, for electrons will easily destroy insects that are ordinarily hard to kill.

Besides the treatment of wheat, the method may be of value in the treatment of rice and cottonseed which may undergo microbiological spoilage during storage. At the present time, there is no satisfactory method of preventing this spoilage.

It should be pointed out that treatment of seeds with cathode rays may destroy the fertility of the seeds, so that the method would not be applicable in those instances where the seed material is to be planted. Data from the Southern Regional Research Laboratory of this Bureau have shown that irradiated cottonseed will germinate even after dosages of 500,000 rep. Data from other laboratories have shown that plants grown from irradiated seeds may show mutations (24).

Fresh Fruits and Vegetables. Citrus fruits are subject to various rots after harvest, depending upon climatic conditions where the fruit is grown and handling and storage methods used.

Blue mold and green mold rots, or penicillium rots of citrus, are very widespread forms of decay during the winter months. Such rots, including stem-end rot will average about 1% annually in shipments, two-thirds of which will be blue mold. Antiseptic dips such as 5% borax solution; sodium orthophenylphenate applied in conjunction with wax emulsion, diphenyl-impregnated fruit wraps, diphenyl-impregnated case liners, and diphenyl-treated separators between the layers of fruit in the shipping box and fumigation with nitrogen trichloride are all said to be effective in checking these rots. However, the chemicals have certain disadvantages, and if cathode ray treatment would prevent such rots, it would have an advantage over chemical treatment. In the treatment of such rots 1 to 2 mm. penetration would be necessary, a depth attainable by present machines. It should be pointed out that some means would be necessary to turn the fruit during treatment so that all parts would be exposed.

Stem-end rot of citrus is less widespread than the penicillium rots, but in the Gulf Coast region it probably causes greater financial losses, and may amount to over $600,000 annually. It is a serious problem in all varieties in Florida and the West Indies where the climate is humid. The condition is combatted with the same chemicals as used for penicillium rots, and recently dips of 2-amino-pyridine have been found successful, but this chemical has not been cleared by the Federal Food and Drug Administration. The effect of cathode rays on the fungi causing stem-end rot would be of interest. However, a penetration of about 1/4 inch would be necessary, which would require a little more than 2 Mev electrons.

Blue mold is probably the most common and most destructive of all rotting organisms that attack apples and pears. Destruction of apples from all decays probably amounts to around 2.9% annually, while blue mold alone accounts for about 1.9%. Control methods have been attempted with washes containing sodium chlororthophenylphenol, but results have not been too encouraging. Irradiation of the fruit with cathode rays may find a place here. Such treatment would require a penetration of 1 to 2 mm.

Attempts have been made to control brown rot in peaches by chlorine dips, but results have been discouraging. Results of treatment with cathode rays would be of interest.

Bacterial soft rot is an important disease of potatoes in storage and in transit. The loss varies from 0.5 to 2.2% annually. It progresses rapidly under some conditions and potatoes that appear to be almost free of the disease when loaded have been known to show 50 to 75% damage after only seven days in transit. Control has not been satisfactory, and it would be of interest to learn the effect of irradiation with cathode rays. Penetration of from 1-2 mm. would probably be necessary.

Prepackaged fresh vegetables, such as spinach, cold slaw, shelled peas, and lima beans, have become of increasing importance during recent years to eliminate preparation by the housewife. However, the life of these commodities is limited, chiefly because of bacterial soft rot, which is particularly serious in the case of shelled lima beans. Irradiation with cathode rays may be of value in overcoming this difficulty, and samples of spinach treated in this manner were seen by representatives of this Bureau. The samples appeared fresh and edible in comparison with the control which was spoiled. It was implied that both the treated sample and control had been stored at room temperature.

Melons (cantaloupe and Persian melons) are usually picked green for shipment, with the result that eating quality will vary greatly when purchased by the consumer. If melons could be picked at their optimum maturity, surface treated with cathode rays, and then shipped, the market for melons would probably be expanded when the consumer learned he could obtain a product of uniform quality Cherries and blueberries packed in consumer containers, sealed with cellophane and then irradiated, are said to keep at room temperature for several weeks. Such products were shown to representatives of this Bureau, and they appeared to be in good condition in comparison to the control.

There is no successful method of preserving corn on the cob; even the frozen product leaves much to be desired. If corn on the cob could be successfully preserved by irradiation, it is obvious that the market for sweet corn would be greatly expanded.

Canned Fruits and Vegetables. Present methods of canning fruits and vegetables all involve heat treatment of different degrees, higher temperatures being necessary for vegetables than for fruits. These temperatures bring about changes in color, flavor, taste and nutritive value in varying degrees depending upon the temperature of processing. Several laboratories are investigating

the use of certain antibiotics in the canning of vegetables for the purpose of using lower temperatures during processing. The possibility of the use of cathode rays in the canning of vegetables and certain fruits would be a subject worthy of investigation. It is not likely that packing could be in tin containers of present commercial size, but it is conceivable that the product could be packed in latex bags (such as Cry-O-Rap) and protected by an outer carton of chipboard. It may also be possible to use plastic containers. This would not only be a saving of tin and steel, but a reduction in shipping costs. Probably not all products could be packed in this manner because of certain oxidative changes brought about by air.

Frozen Foods. Frozen foods are not sterile as are canned foods. Freezing will not destroy microorganisms in the food. Frozen foods have, however, been remarkably free from suspicion of being health hazards both as to enterotoxic food poisoning and infectious diseases. There are several reasons for this, but these will not be discussed here. Treatment of precooked frozen foods with cathode rays would be of interest, because in this type of product the material is not heated sufficiently high prior to consumption to destroy pathogenic organisms should they be present. Packaging would not be such a problem here as in the case of canned foods.

Poultry and Poultry Products. Newcastle disease is an acute, febrile, and contagious virus disease of fowls resembling fowl plague. Because of the prevalence of this disease in certain poultry producing areas of the United States, export of slaughtered poultry to England is not possible. The virus of the disease is centered in the bones, and the disease may be spread by discarding the bones after the consumer is through with the bird. Cooking the poultry, e.g., frying in fat will not destroy the virus in the bones.* Cathode ray treatment would have to be sufficiently deep to reach the bones.

*There has been no record of infection of humans by the consumption of poultry having Newcastle disease. There have been reports of infection of laboratory workers working directly with the virus. Infection was manifested by a conjunctivitis.

In chickens, this would require penetration of about one inch, and in turkeys about 1-1/2 inches. Furthermore, virus destruction requires very high rep.

The life of cut-up poultry under cool storage is comparatively short. If surface sterilization of such poultry could be accomplished by irradiation so that cool storage life could be prolonged to 21 to 30 days, eviscerating plants could service stores over a larger area than is now possible.

Numerous publications have shown that eggs can act as a carrier for Salmonella organisms. Ingestion of bacteria of this group may cause an infection and symptoms in man, similar to, but milder than, typhoid fever. Dried egg albumen, because of its beating quality, is used in certain whips and types of confection which may not be heated sufficiently to destroy Salmonella should they be present in the egg albumen. At present there is no satisfactory method of sterilizing egg albumen without damaging its beating quality. Cathode ray sterilization of egg albumen would be an important contribution to the egg drying industry. Whole liquid eggs can be pasteurized prior to drying or freezing to destroy Salmonella, but this has certain disadvantages, one of which is the fact that pasteurization may harm functions of the eggs important in baking.

Shell eggs are treated with oil to prolong storage life, and attempts have been made to coat the shell with plastics and other preparations, the purpose of both treatments being to prevent loss of moisture. A far more serious problem is the growth of bacteria within the egg. It has repeatedly been demonstrated that the contents of fresh, clean eggs are usually sterile, and that bacterial spoilage of eggs in storage results from penetration of the shell by the organisms after laying. The most important cause of such invasion is undoubtedly the washing of dirty eggs, but it is also likely that the "sweating" of eggs when brought out of a cold room is another potent cause of invasion. Cathode ray treatment along with oiling may be of value in prolonging

storage life. Deep penetration would be necessary, because organisms are chiefly centered in the yolk.

Meat. Today a great many fresh cuts of meat are prepackaged in transparent containers and displayed at ordinary temperatures for selection by the consumer. Shelf life of such meat is very limited. If irradiation would prolong the storage life, it would tend to decrease distribution costs with, perhaps, a saving to the consumer.

The effect of irradiation on the organism causing Rinderpest, and on the virus of hoof-and-mouth disease, may prove of value in the control of these economic crippling diseases. Present regulations, however, would not permit such work to be carried on in the United States.

Dairy Products. Although whole fluid milk is being canned today, the product has not been universally judged satisfactory. A highly acceptable canned whole fluid milk would not only be of importance to the military, but to civilians in remote regions where it is now expensive to ship whole milk and where dry milk must be used.

Evaporated milk is a common market commodity. Its taste, however, is objectionable to most people. This taste is caused chiefly by the high temperatures required for sterilization. If irradiation could be used for sterilization in place of heat, there is a possibility the product could be improved with resulting greater distribution.

Considerable work would probably have to be done on a suitable container for both fluid milk and evaporated milk. Tin containers of present commercial size could not be irradiated because of limited penetration; cardboard containers would probably not be satisfactory because of air penetration and possible development of oxidized and tallowy flavors.

Concentrated milk is now on the market in certain areas of the United States. Its storage life is limited to a few weeks in the household refrigerator. The effect of cathode rays on prolonging the storage life of this product would be an important line of research.

Per capita consumption of cheese in the United States has increased from 4.6 pounds in 1930 to about 7.2 pounds in 1949. Cheese is prone to develop surface mold growth, and propionates are often used as inhibitors, either by immersing the cheese in a solution of the propionate or by dipping the containers, caps or wrappers covering the cheese in the solution.

If surface sterilization of cheese could be accomplished by irradiation rather than by chemical means, the method should be of considerable value to the cheese industry.

Wrappers impregnated with propionates have been used to prevent mold growth on butter, but the practice has not found widespread favor because it gives the butter a propionate odor. Cathode ray treatment may be of value here.

Plant Quarantine. Considerable quantities of fruits and vegetables, plants and seeds are imported into the United States each year. Such material may carry injurious insects and plant diseases and is, therefore, subject to inspection. In a great many instances, plants are imported in crates containing several hundred pounds of dirt, peat and moss. Inasmuch as these crates may be several feet on a side, it would not be possible to treat the contents with cathode rays. Furthermore, such treatment, even if possible, would probably destroy the plants, or, at least, injure them.

Fruits that may be infected with insects, such as the Mediterranean fruit fly or Mexican white fly, could be treated only by penetrating cathode rays; such penetration would have to be of the magnitude of several inches. There are no machines yet available to give such penetration.

In the case of bulbs and seeds, treatment with cathode rays may destroy

the fertility of the material, or, if fertility was not destroyed, the plant may be entirely different from the parent.

It is believed that cathode rays would have no place in the treatment of infected plant material imported into the United States or in the interstate shipment of such plant material.

It is again pointed out that irradiation could not be applied in any of the above cases should unfavorable changes be brought about in the physical appearance of the product or if chemical changes occurred which would adversely affect the nutritive value of the treated food. There is at present no conclusive data that unfavorable changes may not be brought about.

MOST LIKELY FIRST APPLICATIONS

The first commercial application of electron treatment will probably be pharmaceuticals such as antibiotics, sutures, certain vaccines, and antitoxins, chiefly to rid them of pyrogens. All of these biologicals are high in price, and cost of irradiation is not so important as with foods which are cheap in comparison.

Also of probable immediate application is the preservation of human tissue such as blood vessels, bone and cartilage in the establishment of tissue banks. Frozen, irradiated human aortic grafts have been used with apparent success in two humans with coarctation of the aorta, to bridge the aortic gap (25).

In the more distant future, we may expect the application to food materials. Foods first treated would be those of greatest worth per pound, such as meats, poultry, eggs, milk, and certain grains. But before such foods could be offered to the public it will have to be proved that irradiation causes no unfavorable change.

Other possible applications, perhaps still further in the future, are destruction of organisms on cotton; initiation or conducting of chemical reactions such as bodying of drying oils, polymerization of monomers, refining

of glyceride oils to remove color bodies, oxidation of terpenes, addition reactions of olefins, treatment of resin, and certain unsaturated fatty acids, modification of proteins.

PATENTS

There are several patents embodying not only the apparatus used for bombarding with electrons, but methods of treating certain materials to bring about specific chemical or physical changes. It is not the purpose here to discuss the validity of these patents.

In 1936, Brasch and Lange patented a laminated high voltage vacuum discharge tube (26) and in 1937 they received a patent on "an apparatus for producing extremely fast corpuscular rays" (27). In reality, the latter is the capacitron of the Electronized Chemicals Corporation.

In 1947, Brasch (28) obtained patent with 43 claims for irradiation devices for use on an industrial scale, these devices comprising an accelerator tube powered by a Van de Graaff generator, a high-voltage transformer, an impulse generator or "an electrical accelerator to accelerate the velocity of the electrons to a high speed." This appears to be a basic patent.

A method of sterilizing substances by bombarding with high-speed electrons of a velocity equivalent to more than one million volts during a very short time period was patented in 1948 (29). There are patents to cover the aging of alcoholic beverages by bombardment with high-speed electrons (30); methods of making "scenting essential oils" by electronic bombardment of cut plants (31). Another patent deals with the conversion of petroleum fractions (32) or of alcohols (33) to butadiene by repeated bombardment with high speed electrons. A more recent patent (34) claims the destruction of the alkaloids in coffee, tea, and tobacco by high-speed electron bombardment.

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